

# Extrusion and characterization of nanoclay filled polypropylene

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**ABSTRACT:** In recent years the interest in nanocomposites grew exponentially. However, the dispersion of a polar nanoclay in an apolar matrix remains the key issue. In this study, the process factors affecting the dispersion of nanoclay in a polypropylene matrix are studied, being the results obtained in both twin screw and single screw extruders compared. The tensile properties of the material are influenced by the dispersion. Thus, this characteristic is used to evaluate the processing parameters. The main goal of this study is to optimize the extrusion process parameters. The study of the influence of the screw configuration will be done in a future work.

## 1 INTRODUCTION

The interest in nanocomposites grew exponentially during the last decade. Numerous studies show the great potential due to nano-enhanced properties such as improved strength, better fire retardancy and lower gas permeability (Hussain, Hojjati et al. 2006; Paul and Robeson 2008; Kumar, Depan et al. 2009; Pfaendner 2010).

This research focuses on the processing of nanoclay filled polypropylene (PP) using twin screw extrusion. These polymer-layered silicate nanocomposites still struggle to reach the optimal dispersion of the individual polar clay platelets in an apolar matrix, such as polyolefins. The final dispersion of nanoclays in a polymer matrix can be divided in three different composite types: tactoids, intercalated and exfoliated, the latter being the most desired (Pavlidou and Papaspyrides 2008).

In this paper, a general extrusion line for the production of nanocomposites is being proposed. The exfoliation/intercalation of the nanoclay is followed by comparing the Young modulus of the different materials. It is proven that the exfoliation/intercalation of the clay platelets affects the E-modulus of the compounds in a positive way (Lee, Fasulo et al. 2005; Cauvin, Kondo et al. 2010). For a first study, this is a straightforward and easy way to compare the different compounds.

## 2 MATERIALS AND METHODS

### 2.1 Materials

The selected polyolefin was a polypropylene homopolymer PP 531P delivered by Sabic with a MFI of 0,30 g/10min (230 °C/2,16 kg) and an elongation at break of 600%. Cloisite 15A, delivered by Rockwood Clay Additives, was chosen as a montmorillonite based clay, modified with a quaternary ammonium salt (2M2HT) to improve the interlayer distance.

Priex 20070 and Priex 20095, delivered by Addcomp Holland, were used as compatibilizer agents to positively influence the interaction between the apolar polypropylene and hydrophilic organomodified clay. Both are polypropylene-grafted-maleic anhydride (PP-g-MA) with a grafting level of 0,10 wt% and 0,45 wt%, respectively. The higher the level of grafting, the more exfoliation can be reached (Santos, Liberman et al. 2009).

### 2.2 Extrusion machinery/ moulds

A co-rotating twin screw extruder ZSK 18 MEGAlab from Coperion was used to produce the PP nanoclay composites. The temperature settings from hopper to die were 175, 185, 185, 190, 195, 195, 195 and 205°C, respectively. This temperature profile was set as low as possible to avoid clay degradation as experienced in earlier tests and mentioned in the literature as well (Cervantes-Uc, Cauch-Rodriguez et al. 2007).

The PP granules, coupling agent, and clay were metered in the required proportions by using volumetric feeding units. All the materials were dried at 80°C during 6 hours in a vacuum dryer before melt processing.

The twin screw extruder was equipped with a slit die, as shown in figure 1. This die was designed to imitate the continuous large-scale plate extrusion. Because of the low pressure built up (max. 50 bar at head), a small die of 19x2 mm was used.

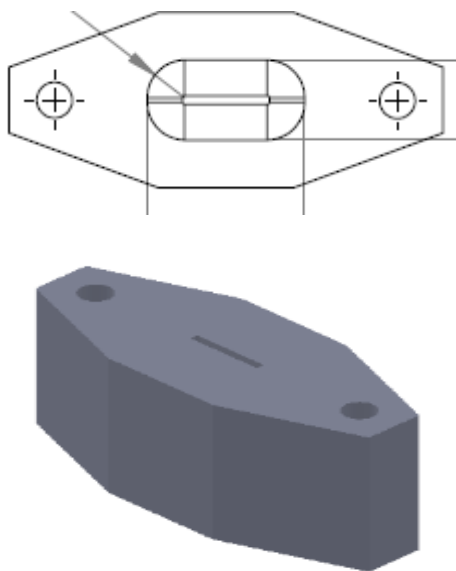


Figure 1. Extrudate slit die (19x2mm)

The screw configuration (figure 2) consists of 10 different zones, with 3 degassing areas and the side feeder mounted on the 4<sup>th</sup> zone. Also present are wide kneading blocks and distribution/mixing elements. This is a standard twin screw set-up with the possibility of being altered. The outer diameter of the screw is 18,1 mm.

Two different screw speeds were tested, as screw speed affects both the output and the residence time of the extruder. The total residence time in the extruder (hopper – die) is 49 and 33 seconds for 200 and 300 rpm, respectively.

The residence time from the side feeder until the die (clay residence time) is 33 and 29 seconds, respectively. The PP feed rate was adjusted at each screw speed to ensure a fully filled screw and an uniform delivery of nanoclay particles.

In the case of the single extrusion tests, an extruder from Brabender Plasti-Corder was used for producing the PP nanoclay composites. The temperature settings from hopper to die were 175, 185, 190, 195 and 205°C, respectively.

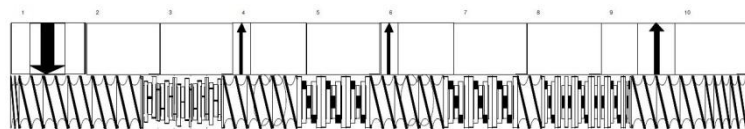


Figure 2. Twin screw configuration

The L/D of the single screw extruder was 15, while for the twin screw extruder, the L/D was 24. The compounds were produced by regranulating the twin screw extrudates and adding them to the single screw extruder.

### 2.3 Sample denotation

Different compounds were produced using both single and twin screw extruders with three different clay concentrations and three different compatibilizer contents. The samples are denoted with PP/compatibilizer/Cloisite 15A followed by the different weight percentages.

The difference between single and twin screw extrusion is denoted as SE (single extruder) and TSE (twin screw extruder).

### 2.4 Characterization

The tensile properties (ISO 527) were measured four weeks after the production of the specimens using an Instron 5565 dynamometer at a tensile speed of 50mm/min. The specimens were fully crystallized and no effects of post-crystallization could intervene. The elastic modulus was calculated automatically by the Blue Hill software. Tensile property values reported in this paper represent an average from measurements on at least five specimens.

The crystallinity of the specimens was determined using a Netzsch DSC 204F1 at a heating rate of 10°C/min under nitrogen atmosphere. The crystallinity of the different compounds was calculated as follow:

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where  $\Delta H$  is the apparent enthalpy of fusion per gram of composite,  $\Delta H^0$  is the heat of fusion of a 100% crystalline PP, and  $w$  is the weight fraction of organoclay in the specimens.

### 3 RESULTS AND DISCUSSION

First the results of the different processing parameters for the twin screw extruder are discussed in sections 3.1, 3.2 and 3.3. Afterwards (section 3.4), a comparison between twin and single screw extruders is shown.

#### 3.1 Influence of the screw speed and temperature profile

The screw speed of the extruder affects the output and the residence time in the extruder. A higher residence time can lead to a higher degree of intercalation/exfoliation of the nanoclay in the PP matrix and hence improve the mechanical properties (Treece, Zhang et al. 2007).

Figure 3 displays the modulus results for different compounds. All samples show a loss in E-modulus for higher screw speeds. When the residence time of the nanoclays in the extruder is higher, the different polymer molecules have more time to penetrate between the interlayers of the organoclays. This intercalation/exfoliation leads to a higher E-modulus of the samples for lower screw speeds.

The E-modulus of the tested virgin PP stays mostly constant around 850MPa for the different screw speeds.

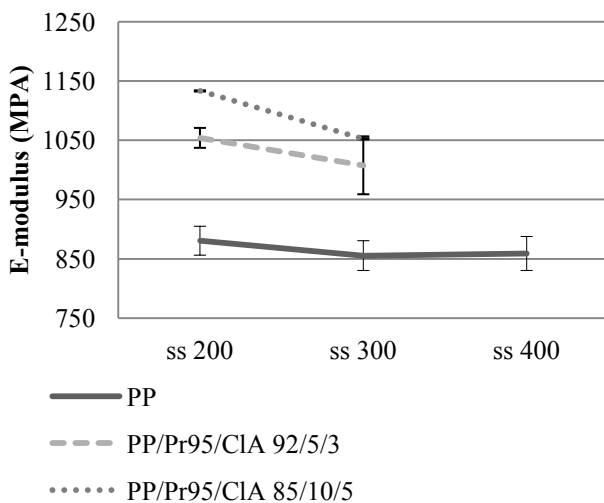


Figure 3. Influence of the screw speed on the E-modulus of the material (resp. 200, 300 and 400 rpm)

#### 3.2 Influence of the %wt nanoclay with the same amount of compatibilizer

The influence of different percentages nanoclay were tested with the same constant amount of compatibilizer (5 wt%). Figure 4 shows the E-modulus of the compounds with different clay concentrations.

There can be concluded that an optimum is reached with 3 wt% nanoclay.

If the amount of compatibilizer would increase, the modulus of the 5 wt% sample would probably also increase. This is shown in section 3.3. When increasing the clay concentrations, one has to raise the compatibilizer content as well.

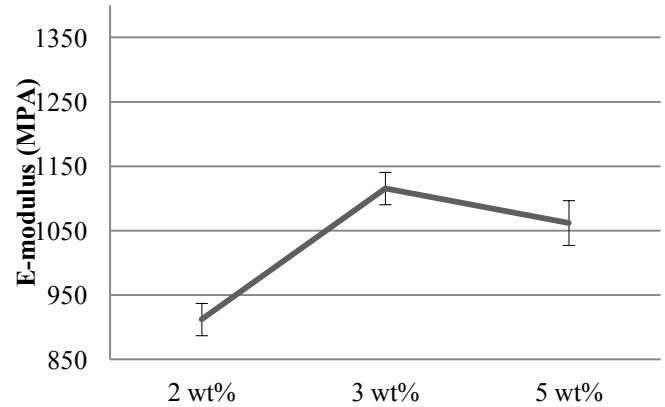


Figure 4. Influence of %wt cloisite 15A on the E-modulus of the material (5% compatibilizer)

#### 3.3 Influence of the %wt compatibilizer

Figure 5 illustrates the E-modulus with different %wt compatibilizer with the same amount (5 wt%) of Cloisite 15A. The screw speed was kept constant at 200 rpm. The higher the amount of coupling agent, the higher the free maleic anhydride concentration, leading to a better compatibility between the apolar PP and the polar clay platelets. This is shown below with a constant raise (at low concentrations) of the elastic modulus.

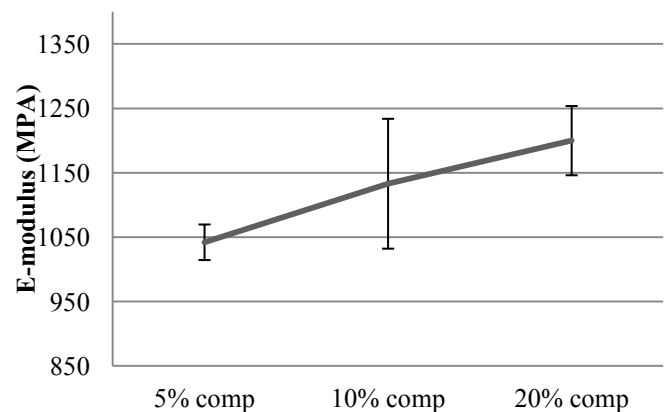


Figure 5. Influence of the %wt compatibilizer (Priex 20095) on the E-modulus of the material (5 wt% Cloisite15A)

### 3.4 Comparison between twin and single screw extrusion

Twin screw extrusion is normally used to produce nanocomposites due to the higher mixing capabilities in comparison with single screw. Single screw extrusion leads to nanocomposites with lower E-modulus as can be seen in figure 6. A lower mixing capacity leads to a loss in penetration possibilities for the polymeric chains into the organoclays and hence lowers the intercalation/exfoliation.

In figure 6, two runs are displayed, to indicate the trend that was being noticed. The standard deviations of the different measurements are not mentioned because of the overlap, but are in the order of 30 MPa for each case.

The E-modulus of the samples drops both to approximately the same value.

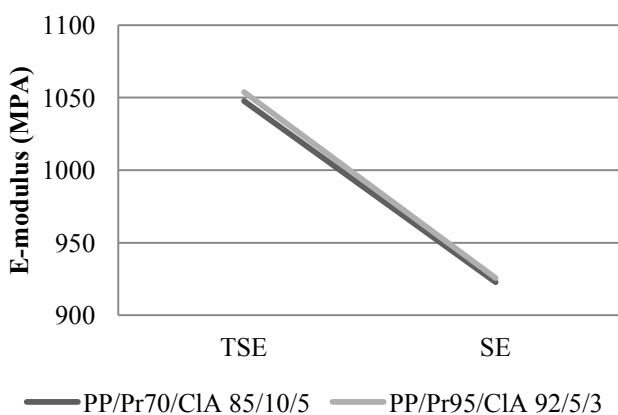


Figure 6. Comparison between twin screw extrusion and single screw extrusion

### 3.5 Degree of crystallization

Table 1: Crystallinity

|             | wt%    | Cryst% |
|-------------|--------|--------|
| PP          | 100    | 40     |
| PP/Pr95     | 90/10  | 39     |
| PP/Pr95/CIA | 92/5/3 | 43     |
| PP/Pr95/CIA | 90/5/5 | 43     |

The incorporation of clay into the PP matrix induces a minor increase in crystallinity. The virgin PP with the coupling agent reduces the crystallinity (table 1). These results were tested with different clay concentrations and different compatibilizers.

## 4 CONCLUSIONS AND FUTURE WORK

Different conclusions can be drawn from these results:

- A lower screw speed (TSE) gives stiffer nanoclay filled polypropylene with higher E-modulus.
- When increasing the clay content, one has to increase the compatibilizer concentration as well.
- Twin screw extrusion is more suited for the production of nanoclay filled polypropylene, due to the higher mixing capacity and hence better dispersion and higher<sup>2</sup> Young modulus of the samples.

Additionally, too high concentrations of organoclay (without increasing the compatibilizer content) gives brittle materials. Adding some rubber particles to the polymer matrix could lower this problem.

The crystallinity of the samples is only slightly influenced by adding nanoclay particles.

The current screw set-up allows us to produce nanoclay filled polypropylene with an increase in elastic modulus up to 35 %. This research gives us, based on these results a good start to influence the screw configuration and compare them with current results.

Future work will include visualisation techniques as SEM/TEM and XRD to ensure the correct dispersion of the nanoclay in the polypropylene. The other focus will be on the screw modification, as in the present paper only a standard twin screw was used. By changing the screw configuration, different shear rates can be achieved and compounds with different properties can be produced.

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